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MAGNETO-DIELECTRIC PROPERTIES OF TRANSFORMER OIL BASED MAGNETIC FLUIDS IN THE FREQUENCY RANGE UP TO 2 MHz

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We have studied dielectric and magnetodielectric properties of transformer oil UTR 40 based magnetic fluid with various concentrations of magnetic nanoparticles of Fe_3O_4 covered with oleic acid as a surfactant. The experiments were conducted at different volume concentrations of magnetite nanoparticles at room temperature and in a frequency range from 100 Hz up to 2 MHz with and without external magnetic field up to 30 mT. The quasilinear increase of the dielectric constant with volume concentration has been confirmed. The variation in dielectric permittivity with frequency reveals that the dispersion exhibited by the samples is due to a Maxwell–Wagner type interfacial polarization. The dielectric anisotropy factor $g(B,\omega)$ is very close to $g = 1$.

Introduction. Magnetic liquids or ferrofluids are new technological materials which are of great interest for applications. The physical properties of these liquids determine many applications in different fields such as sealing and grinding technologies, heat transfer and hydrodynamic flow applications. Investigation of dielectric and magnetodielectric properties of magnetic fluids is intimately connected with the magnetodielectric effect that is characterized by magnetodielectric anisotropy. The dielectric behaviour of magnetic fluids changes with the application of an external magnetic field and with the relative orientation of electric and magnetic fields. This effect is known as the magnetodielectric anisotropy effect. Magnetodielectric effects in magnetic fluids have been investigated by many researchers [1]–[6] both experimentally and theoretically. The experimental investigations were based on impedance measurement techniques, where the magnetic fluid was placed in a capacitor. Measurements of the impedance parameters such as the modulus and phase are carried out using a bridge or an RLC meter. It is known that the impedance measurement techniques suffer from some serious disadvantages such as electrode effects, parasitic impedances, skin depth, and accuracy related problems. Very recently Yusuf *et al.* [7] have determined the magnetodielectric effect from magnetooptical measurements, where the disadvantages inherent to conventional impedance measurement techniques are avoided. When the magnetic fluid is exposed to a magnetic field, the processes in macroscopic surroundings of the electrode system and microscopic surroundings of magnetic particles create needle-shape aggregations (clusters) of magnetite particles. One of the ways to resolve these processes is an equation that expresses the effect of force action of magnetic and electric fields in the surroundings. The force causing the mobility of particle clusters in the electric field is dependent on their weight that is dependent on the volume concentration of magnetite particles and local density of magnetic fluids. The Stokes force, which expresses the effect of dynamic viscosity of magnetic fluids, is not eligible at application of the alternating electric field with the frequency 50 Hz. Many authors focused their research on the influence of external magnetic and electric fields on the dielectric properties of magnetic fluid at lower frequency values. Ferrites are suitable for application in a circuit with a frequency range above MHz as a consequence of the high value of specific electrical resistivity ($10 - 10^4$ Ωm) and small eddy currents [8]. Very important is that the dielectric losses in this MHz frequency range are very low that predominates the low value of saturation magnetization. The use of ferrites at higher frequencies requires the knowledge of the dynamic properties at higher frequencies.

The goal of the work is to give a proof about the real course of frequency dependencies of both the relative permittivity and the loss factor of magnetic fluids based on transformer oil UTR 40 with different concentrations of magnetic particles and magnetodielectric effect in the measured frequency range up to 2 MHz.

1. Theory. When alternating voltage acts at steady state, a time-varying electric field has to be taken into consideration at stress of a liquid medium. It means that the characteristic electric quantities are dependent on the frequency. The specific electric conductivity is not dependent on the frequency in steady state if electric charge carriers during half-period do not travel a trajectory comparable with their mean free trajectory in the observed medium. If that requirement is fulfilled, then the specific electric conductivity decreases with the increase of frequency and the conductivity mechanism converts to the state of relaxation mechanism. The relaxation processes are frequency dependent. This influence will manifest itself strong if the relaxation time constant is comparable with the period of alternating voltage. As a consequence, both the relative permittivity and the loss factor tgα of the observed medium will be frequency dependent.

It is necessary to take into account the total polarization of the liquid medium to derive the frequency dependence of relative permittivity. It expresses the polarizability effectiveness of k particles of i number of particles with a concentration of n_i. If the size of the electric dipole moment is m_i and E is the size of applied electric field intensity, then

$$m_i = a_i \cdot E, \tag{1}$$

where a_i is the polarizability of particles. The intensity of the electric field in an immediate vicinity of particles is not the same as for a macroscopic applied electric field. So the total polarization will be given as

$$P = \sum_{i=1}^{k} n_i m_i, \tag{2}$$

and the equation expressing the frequency dependence of relative permittivity will have the form

$$\epsilon_\gamma = 1 + \frac{1}{\epsilon_0} \sum_{i=1}^{k} \frac{n_i a_i}{1 + \omega^2 \tau_i^2}, \tag{3}$$

where ϵ_0 is the vacuum permittivity, n_i is the particles concentration, m_i is the electric dipole moment of particles, $\omega = 2\pi f$ is the angular frequency of the applied electric field of sinusoidal shape, τ_i is the time constant, a_i denotes polarizability.

The equations for the frequency dependence of volume losses γ and loss factor $\tan\delta$ can be derived as [9]

$$\gamma = \gamma_0 + \sum_{i=1}^{k} n_i a_i \frac{\omega^2 \tau_i}{1+\omega^2\tau_i^2}, \tag{4}$$

$$\tan\delta = \frac{\gamma_0 + \sum_{i=1}^{k} n_i a_i \frac{\omega^2\tau_i}{1+\omega^2\tau_i^2}}{\omega\epsilon_0 + \sum_{i=1}^{k} n_i a_i \frac{\omega}{1+\omega^2\tau_i^2}}. \tag{5}$$

The goal of the work is to give a proof about the real course of frequency dependencies of both the relative permittivity and the loss factor of magnetic fluids based on transformer oil UTR 40 with different concentrations of magnetic particles.

2. Experiment. The used magnetic fluid (MF) was produced based on transformer oil UTR 40 and magnetite (Fe_3O_4) nanoparticles, with oleic acid as a surfactant. The synthesis of a MF with an organic liquid carrier based on the established procedure [10] followed these main steps: synthesis of surface coated magnetite nanoparticles: co-precipitation (at 80°C) of magnetite from aqueous solutions of Fe^{3+} and Fe^{2+} ions in the presence of a concentrated NH_4OH solution (25%, sterical stabilization (chemisorption of oleic acid; 80–82°C), magnetic decantation and repeated washing, extraction of monolayer covered magnetite nanoparticles (acetone added; extraction) to obtain stabilized magnetite nanoparticles. Then, the magnetic nanoparticles were dispersed in a non-polar liquid carrier at 120–130°C followed by magnetic decantation/ filtration, flocculation and re-dispersion of magnetic nanoparticles to produce a non-polar magnetic nanofluid. The magnetization curves of the samples were measured by vibrating sample magnetometry using a VSM magnetometer (Model 880, DMS/ADE Technologies, USA) at room temperature ($\approx$25°C) in a magnetic field up to 800 kA/m. The measuring plan-parallel cupreous electrodes were placed into a Faraday cage to measure dielectric parameters of magnetic fluids with a possibility to insert permanents magnets for measurements in a magnetic field (Fig. 1). The electrodes were 1 cm in radius and the distance between the electrodes was 0.5 mm. The device for capacity and loss factor measurement was connected to a microcomputer and to a generator of signal with a varying frequency (100 Hz − 2 MHz) so that the output voltage was 2 V. The measurements were carried out at a temperature of 20°C. The relative permittivity was measured using the Shering bridge Tettex 2818. The experimental error of the capacity measurements was 0.05% and of the loss factor 1%. The relative permittivity of magnetic liquids was determined from the capacity measurements as $\epsilon_r = C/C_0$, where C is the capacity of a capacitor with a magnetic liquid as a dielectric and C_0 is the capacity of the same capacitor filled with air. Two permanent NdFeB magnets were used as a source of a uniform magnetic field up to 30 mT. The capacity and loss factor have been measured at both parallel and perpendicular mutual orientations of the electric and magnetic fields and

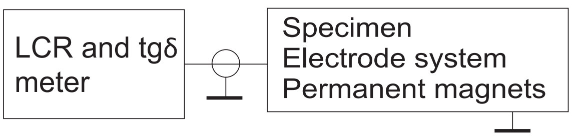

Fig. 1. Schematic presentation of the experiment.

without magnetic field. The magnetic fluids based on transformer oil with various concentrations of magnetite nanoparticles up to 2.03% magnetic volume fraction and on pure transformer oil were used in our experiments. The magnetic volume fraction was obtained from the value of saturation magnetization, M_{s}, assuming the saturation magnetization for solid magnetite.

3. Results and discussion. In our experiment, we have investigated five samples of magnetic nanofluid, with the liquid transformer oil UTR 40 carrier and magnetite nanoparticles as the magnetic dispersed phase produced by the above-mentioned method. The five samples of different volume fractions of magnetite were prepared by dilution of the most concentrated one with the carrier liquid MF1, MF2, MF3, MF4 and MF5 with saturation magnetization 15, 35, 62, 97 and 114 Gauss, respectively. The magnetic volume fraction was obtained from the value of saturation magnetization, M_{s}, assuming the saturation magnetization for magnetite $M_B = 4.46 \times 10^5$ A/m according the following relation:

$$\phi_{\mathrm{m}} = \frac{M_{\mathrm{s}}}{M_B}. \tag{6}$$

The volume fraction resulted from Eq. (6) using the densities of the magnetic nanofluid, magnetite $\rho_{\mathrm{P}} = 5.18$ g/cm^3 and transformer oil $\rho_{\mathrm{F}} = 0.867$ g/cm^3 is:

$$\phi = \frac{\rho_{\mathrm{MF}} - \rho_{\mathrm{F}}}{\rho_{\mathrm{P}} - \rho_{\mathrm{F}}}. \tag{7}$$

The physical properties of the produced magnetic fluids are given in Table 1.

The dependence of the relalative permittivity on the MF magnetization saturation illustrated in Fig. 2 exhibits a quasilinear behaviour, as can be observed for

Table 1. The basic physical parameters of the produced transformer oil based magnetic fluids.

	M_{s} [Gauss]	ρ [g/cm^3]	Magn.vol. fraction [%]	Vol. fraction [%]
MF1	15	0.901	0.29	1.01
MF2	35	0.937	0.62	1.62
MF3	62	0.960	1.10	2.15
MF4	97	0.996	1.73	2.99
MF5	114	0.998	2.03	3.03

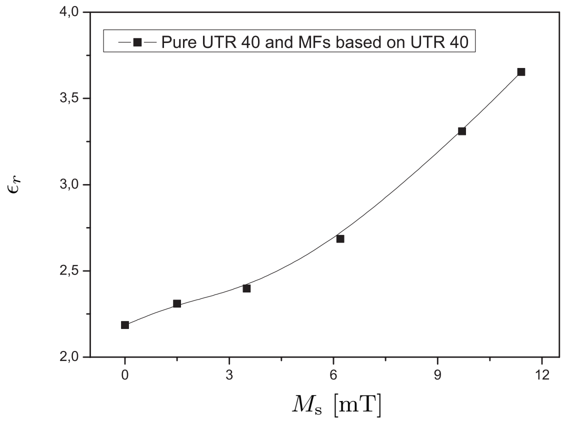

Fig. 2. Dependence of the permittivity on the magnetization saturation corresponding to the magnetite concentration of a transformer oil based magnetic fluid.

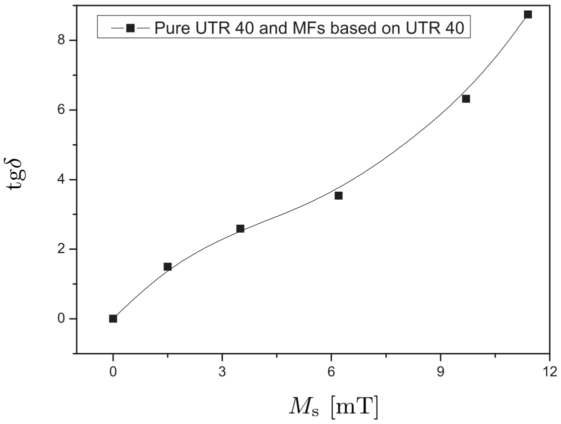

Fig. 3. Dependence of the loss factor on the magnetization saturation corresponding to the magnetite concentration of a transformer oil based magnetic fluid.

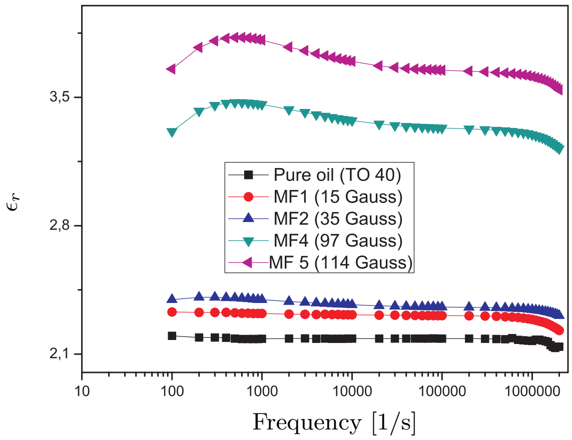

Fig. 4. Dependence of the relative permittivity on the frequency for various magnetic volume concentrations of nanoparticles.

linear dielectrics (see ferroelectric ceramic). The relative permittivity for $M_s = 0$ T belongs to pure UTR 40. A similar behaviour was acquired for the dependence of the loss factor on the MF saturation magnetization (Fig. 3). The observed dependence is caused by the presence of magnetite nanoparticles covered with oleic acid, which have a relatively high specific conductivity (category of semiconductors) that causes the increase of dielectric losses with the increase of MF saturation magnetization. It is very important for magnetic fluids application to observe their dielectric, resp. magneto-dielectric properties dependent on the frequency of an applied electric field. These materials belong to a group of weak polar liquids dependent on the concentration of magnetic particles. It is known that the relative permittivity in magnetic fluids is dependent on a lot of factors as the type of surfactant around the particles, concentration, zeta potencial, pH value and so on [12], and it is not at all easy to generalize the description of the obtained results. Equation (3) describes the ideal dependence of relative permittivity on frequency, but magnetic fluids are more complex colloidal systems. That is why the behaviour of the relative permittivity at low values of MF saturation magnetization corresponds to a theoretical assumption (Fig. 4). The crossing point was

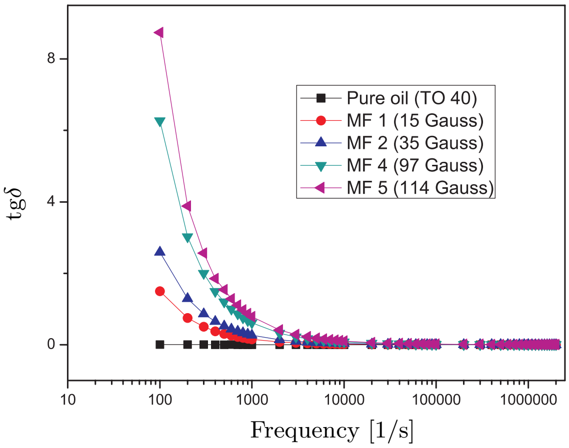

Fig. 5. Dependence of the loss factor on the frequency for various magnetic volume concentrations of nanoparticles.

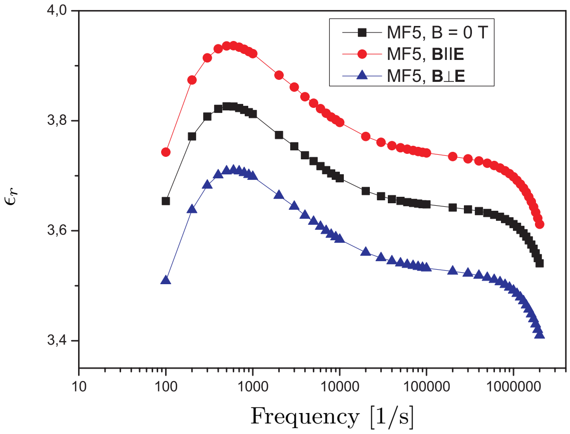

Fig. 6. Frequency dependence of the relative permittivity for MF5 ($\phi = 2.03\%$) in various mutual orientations of electric and magnetic fields.

reached at a frequency of 1 kHz. When the frequency of electric voltage exceeds 1 kHz, the behaviour is again monotonous. It means that a weak orientational polarization appearing at lower values of saturation magnetization (MF1, MF2) of magnetic fluids causes a low and constant relative permittivity. The permittivity for magnetic fluids with a higher saturation magnetization (MF4 and MF5) is not linear dependent on frequency, that is why the orientational polarization in these fluids is not dominated. The frequency crossing point is again at 1 kHz. The variation in dielectric permittivity with frequency reveals that the dispersion exhibited by the samples is due to the Maxwell–Wagner type interfacial polarization and in agreement with Koops phenomenological theory [13], similarly as it was observed in chitosan magnetite nanocomposites [13]. The loss factor tan(δ) is a critical factor for investigation of energetic conditions in magnetic fluids and also for determination of their application possibilities (for example, in transformers of high voltages). A result from Fig. 5 is the fact that electric dipoles of particles are able to follow a change of the alternating electric field at low frequencies so that dielectric losses increase with the increase of MF saturation magnetiza-

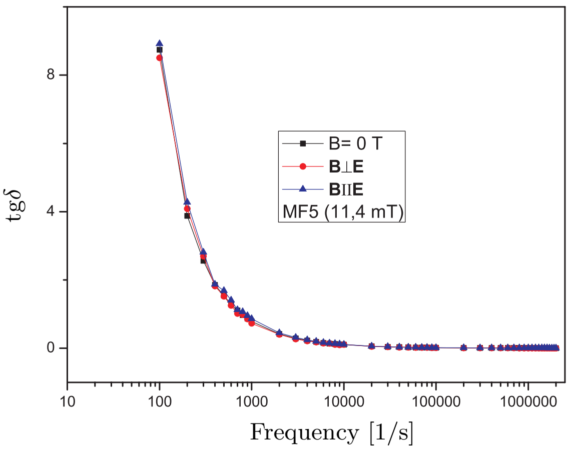

Fig. 7. Frequency dependence of the loss factor MF5 ($\phi = 2.03\%$) for various mutual orientations of external electric and magnetic fields.

tion. The magnetite particles ability to follow the variation of the electric field at high frequencies decreases so that dielectric losses decrease with the increase of the electric voltage frequency. The dependence of the relative permittivity on the electric field frequency for a magnetic fluid with the highest value of magnetic volume concentration (a magnetization saturation of 114 Gauss), with the external magnetic field simultaneously applied parallel and perpendicular to the electric field, is shown in Fig. 6. When a continuous magnetic field of 30 mT is applied, the permittivity of the magnetic fluid increases for parallel and decreases for perpendicular orientation for the whole range of the used frequency with the same character of dependence. The induced anisotropy in relative permittivity of a magnetic fluid subjected to an external magnetic field can be observed. The magnetodielectric anisotropy factor g vs. frequency is very close to $g = 1$ in all frequency interval that is in agreement with previously published results [1]. The variation of the dielectric permittivity with the applied magnetic field can be resulted from an alignment of non-interacting nanoparticles as a consequence of the coupling between easy axes and the magnetic moment of particles. The similar dependence of dielectric anisotropy was observed for other concentrations as well. The anisotropy characterized by deviation of permittivity in individual mutual orientations of electric and magnetic fields showed that the highest value was obtained for the highest volume concentration. The obtained results correspond to the results obtained in [11], where the magnetodielectric effect on the volume concentration and intensity of the electric field were studied. As shown in Fig. 7, the loss factor measured in the presence of an external magnetic field has the same character as for the relative permittivity (Fig. 7), i.e. the values of the MF loss factor increase for parallel and decrease for perpendicular orientation in all frequency range.

Conclusion. In this work, we have described the experimental results on the relative permittivity and loss factor of transformer oil based magnetic fluids with nearly spherical magnetite nanoparticles with the magnetite volume fraction up to 2.03% measured in the frequency range from 100 Hz to 2 MHz. The value of the relative permittivity and loss factor increases almost linearly with the volume concentration of magnetite nanoparticles as a consequence of the higher specific conductivity. The variation in dielectric permittivity with frequency reveals that the dispersion exhibited by the samples is due to the Maxwell–Wagner type interfacial polarization. The frequence dependence of permittivity shows the peculiarities mainly at higher concentrations of magnetite nanoparticles as a maximim at low frequencies (below 1 kHz) and the sharp decrease at 1 MHz. For explanation, more experiments will be needed.

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